8th Int. Symp. "Nanostructures: Physics and Technology"
St Petersburg, Russia, June 19–23, 2000

Light confinement in quantum dots

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Abstract. A concept of light confinement in quantum dots due to diffraction of electromagnetic waves at the dot boundary, is introduced. Possible manifestations of the phenomenon, such as depolarization shift of the exciton frequency, polarization-dependent splitting of the gain band, asymmetry of the absorption and gain spectra, induced magnetization of quantum dots, contribution to radiative lifetime, are discussed both for isolated quantum dots and quantum dot ensembles. We propose that the effect of light confinement should be properly addressed to optimize the design of optoelectronic devices involving quantum dots.

Introduction

A fundamental breakthrough in semiconductor device physics is connected with the recent progress in the synthesis of sheets of nano-scale 3D confined narrow-gap insertions in a host semiconductor, quantum dots (QDs). The large body of recent results on physical properties of QDs and their utilization for the QD laser design has been accumulated in a monograph [1]. The key peculiarity of QDs emerges from the 3D confinement of the charge carriers determined by QD size and shape. However, there exists a class of effects governed by the QD size and shape, which have not received much attention so far. These effects are related to resonant nature of the exciton which provides a dramatic resonant discontinuity of the permittivity at the QD boundary and, consequently, gives rise inhomogeneity of the electromagnetic field both inside and outside QD. By analogy with charge carrier confinement, redistribution of the electromagnetic field energy between the QD interior and exterior under effect of the QD boundary can be referred to as *light confinement*. In many cases the role of light confinement can properly be accounted for the formation in QD of depolarization electromagnetic field, e.g., in dipole approximation of the diffraction theory.

To our knowledge, some physical consequences of the light confinement in an individual QD first time were considered by Schmitt-Rink *et al.* [2]. Manifestation of this phenomenon in relation to the scanning near-field optical microscopy was discussed by Martin *et al.* [3] for geometrically complex mesoscopic systems and by Hanewinkel *et al.* [4] for QDs. An asymmetry of optical absorption and gain spectra in single QD because of depolarization field has been mentioned in Ref. [4]. Recently it has been predicted and experimentally verified that the light confinement in QD arrays constituted by anisotropically shaped QDs manifests itself as polarization splitting of the gain band [5] and, in more general case, as the fine structure of this band [6]. Such a splitting was first experimentally observed by Gammon *et al.* [7], where the depolarization field effect has been proposed as possible

explanation of the splitting. Some new effects related to the light confinement in QDs are considered in Ref. [8].

In our paper we introduce sequential concept of light confinement in 3D-confined resonant nanoinsertions and discuss some general consequences of this phenomenon in an isolated QD and in QD arrays. Our consideration is based on classical electrodynamics of inhomogeneous media.

1. Depolarization shift of the exciton resonance

1.1. Polarizability of a single QD

Conventional phenomenological model of the gain in a QD is based on semi-classical theory of two-level systems which gives the equation of motion for the mean polarization $\mathcal{P}$ caused by transitions between the levels:

$$\left(\frac{\partial^2}{\partial t^2} + \frac{2}{\tau}\frac{\partial}{\partial t} + \omega_0^2\right)\mathcal{P} = -\frac{\omega_0}{2\pi\varepsilon_h}\widehat{\mathbf{g}}_0\,\mathcal{E}\,. \tag{1}$$

Here ω_0 is the exciton resonant frequency and τ is the exciton dephasing time in QD. The phenomenological parameter $\widehat{\mathbf{g}}_0$ is proportional to the oscillator strength of the transition. In anisotropically shaped QDs this parameter is tensorial owing to anisotropy of the charge carrier confinement [10]. In an inverted medium $(\widehat{\mathbf{g}}_0)_{ij} > 0$. The field $\mathcal{E}$ stands for the field inside the QD, different from the external acting field $\mathbf{E}$. This difference is determined by the depolarization field which is as follows [9]: $\mathcal{E} = \mathbf{E} - 4\pi\widehat{\mathbf{N}}\,\mathcal{P}$, with $\widehat{\mathbf{N}}$ as the depolarization tensor. This tensor is symmetrical and depends only on the QD shape. If we neglect the contribution of the depolarization field putting $\mathcal{E} = \mathbf{E}$ into Eq. (1), solution of this equation in the vicinity of resonance for time-periodic fields and isotropic $\widehat{\mathbf{g}}_0 = g_0\widehat{\mathbf{I}}$ gives the well-known Lorentz contribution to the medium polarizability: $\alpha(\omega) = (g_0/\varepsilon_h)[\omega - \omega_0 + i/\tau]^{-1}$, which is commonly used as phenomenological model of the dispersion and the gain of a single QD: $\varepsilon_d(\omega) = \varepsilon_h[1 + \alpha(\omega)]$. Otherwise, taking into account the contribution of the depolarization field, we obtain the tensorial polarizability of QD in the vicinity of resonance:

$$\widehat{\boldsymbol{\alpha}}(\omega) = \frac{1}{\varepsilon_h}\left[\omega\widehat{\mathbf{I}} - \left(\omega_0 - \frac{i}{\tau}\right)\left(\widehat{\mathbf{I}} - \frac{1}{\varepsilon_h\omega_0}\widehat{\mathbf{g}}_0\widehat{\mathbf{N}}\right)\right]^{-1}\widehat{\mathbf{g}}_0\,. \tag{2}$$

Thus, the QD's shape reflects itself as fine structure of the resonance which itself is a superposition of three bands with frequencies $\omega_N^{(j)} = \omega_0 - \nu_j$, $i, j = 1, 2, 3$, where ν_j are the eigenvalues of the inner tensorial product $\widehat{\mathbf{g}}_0\widehat{\mathbf{N}}/\varepsilon_h$. For spherical inclusions the tensors $\widehat{\mathbf{N}}$ and $\widehat{\mathbf{g}}_0$ are isotropic and the fine structure manifests itself as a polarization-independent shift of the gain line depicted in Fig. 1. If the energy splittings are much less than the bandwidth, which means the inequality for energy spacings $\max|\Delta\omega_{ij} = \omega_N^{(i)} - \omega_N^{(j)}| \ll 2/\tau$ to be true, the depolarization field will lead to a distortion of the gain band similar to the inhomogeneous broadening. Otherwise, when $|\Delta\omega_{ij}| \sim 2/\tau$, three separate bands will appear in the gain spectrum of a QD array.

1.2. Birefringence in QD arrays

Since the QD linear extension is much smaller than the resonance wavelength, electromagnetic properties of such ensembles – composite materials – can be modeled in the framework

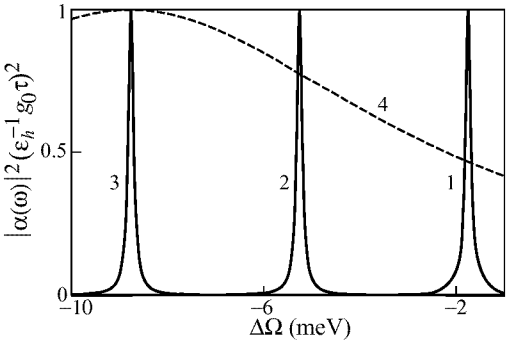

Fig. 1. Depolarization shift of the exciton resonance in spherical QD. Input parameters: $\varepsilon_h = 12.25$, $\lambda = 1\mu$m, $\Delta\Omega = \omega - \omega_0$, $\tau = 10^{-11}$ s, $g_0 = 1 \times 10^{14}$ (1), 2.5×10^{14} (2) 5×10^{14} (3) s^{-1}. For curve 4, $\tau = 10^{-13}$ s, $g_0 = 5 \times 10^{14}$ s^{-1}.

of the effective-medium approach. Thus, a homogeneous medium with effective constitutive parameters instead a composite is being considered. We restrict ourselves to a regular array of QDs arranged in a tetragonal lattice. Assuming QD to have a symmetry axis aligned with the lattice vector $\mathbf{e}_z$, the effective permittivity tensor of the composite can be expressed in terms of a Cartesian basis diadics by $\widehat{\varepsilon}_{\text{eff}}(\omega) = \varepsilon_H(\omega)(\mathbf{e}_x\mathbf{e}_x + \mathbf{e}_y\mathbf{e}_y) + \varepsilon_E(\omega)\mathbf{e}_z\mathbf{e}_z$, where

$$\varepsilon_\sigma(\omega) = \varepsilon_h + \frac{f_V\alpha_\sigma(\omega)}{1 + f_V\delta_\sigma\alpha_\sigma(\omega)}, \tag{3}$$

and $\sigma = (E, H)$ refers to light polarized along (E-polarization) or normal (H-polarization) to the z-axis; f_V is the volume fraction of QDs. The polarizability components α_σ follow from Eq. (2). The depolarization factors N_σ and the geometrical coefficients δ_σ (see [5, 6]) correspond to two different mechanisms responsible for modification of the gain in arrays. The first mechanism is related to the light confinement at individual QDs. The second one is a collective effect defined by electromagnetic interaction between QDs in the ensemble. The combined effect of both mechanisms is given by

$$\omega_N^{(\sigma)} = \omega_0 - \frac{g_0^\sigma}{\varepsilon_h}(N_\sigma + f_V\delta_\sigma)\ , \quad \Gamma_{(\sigma)} = \frac{1}{\tau}\left(1 - \frac{g_0^\sigma}{\varepsilon_h\omega_0}\right). \tag{4}$$

The phenomenological temporal parameter τ in these equations is the collective characteristics of the QD array which must be extracted from the experiment.

In the language of crystal optics, the QD composite being considered is effectively a uniaxial dielectric medium with the z axis as its preferred axis. The phenomenon of birefringence is characteristic for this medium: Both ordinary and extraordinary planewave propagation can occur in it. The refractive indices of these waves, n_H and n_E, respectively, are given by

$$n_H = \sqrt{\varepsilon_H}\,, \qquad n_E = \left[\frac{\varepsilon_E\varepsilon_H}{\varepsilon_H + (\varepsilon_E - \varepsilon_H)\cos^2\theta}\right]^{1/2}, \tag{5}$$

where θ is the angle between the z axis and the propagation direction. Eq. (5) shows that $n_H = n_E$ when the propagation direction coincides with the z axis ($\theta = 0$), and $n_E = \sqrt{\varepsilon_E}$ when the propagation occurs in the x0y plane ($\theta = \pi/2$). Distinction between n_H and n_E in this geometry is responsible for the polarization splitting of the gain band described in details in Refs. [5, 6].

2. Radiative lifetime of spherical QD

Let an isolated spherical QD of the radius R be exposed to an external time-harmonic electromagnetic field. Well-known exact solution of the diffraction problem for a sphere is essentially simplified [11] in view of the condition $kR\sqrt{\epsilon_h} \ll 1$, which is valid for any realistic QDs. This solution and presents the field outside the sphere in terms of its electric and magnetic polarizabilities:

$$\alpha^e(\omega) = \frac{3[\varepsilon_d(\omega)F(\kappa) - \varepsilon_h]}{[\varepsilon_d(\omega)F(\kappa) + 2\varepsilon_h](1 - ikR\sqrt{\varepsilon_h}) + i(kR)^2\varepsilon_h^2 F(\kappa)} \tag{6}$$

$$\alpha^m(\omega) = \frac{3[F(\kappa) - 1]}{[F(\kappa) + 2](1 - ikR) + i(kR)^2 F(\kappa)} . \tag{7}$$

Here $\kappa = kR\sqrt{\varepsilon_d(\omega)}$, $F(\kappa) = (\sin\kappa - \kappa\cos\kappa)/[(\kappa^2 - 1)\sin\kappa + \kappa\cos\kappa]$; $F(\kappa) = 1$ in dipole approximation. In QWs, the problem of the radiative lifetime evaluation is solved by finding of frequency poles of the reflection and transmission coefficients for TE- and TM-polarized plane waves (see, e.g., [12]). Real parts of these poles determine resonant frequencies while imaginary parts give the homogeneous linewidths, which are sums of the dephasing broadening and the radiative broadening. For QDs, we must evaluate the poles of the electric and magnetic polarizabilities. In dipole approximation, simple manipulations lead to

$$\tau_{\text{rad}}^{QD} \simeq -\frac{9}{4\pi^2 g_0}\left(\frac{\lambda}{R}\right)^2 . \tag{8}$$

We note that the material gain g_0 in QD is incorporated as phenomenological parameter in this equation. It is, indeed, a function of the QD size, shape, strain distribution and an effective coefficient of light confinement in a QD. Depending on particular situation g_0 can either increase or decrease with R. In the case when the light confinement is not relevant and the overlap integral is not a function of QD size (the case which is shown to be not correct for real QDs) $g_0 \sim R^{-3}$. In any case, Eq. (8) shows *additional* radial dependence of the radiative lifetime as compared to the conventional dependencies (see, e.g., [13, 1, 14]). Figure 2 presents radiative lifetime numbers obtained from different theoretical approaches and experiments. Conventional model including the realistic overlap integral [14] is shown in Fig. 2 as open squares while this model with unit electron and hole wavefunction overlap integral [13] is presenter by dashed line. The experimental results for radiative lifetimes

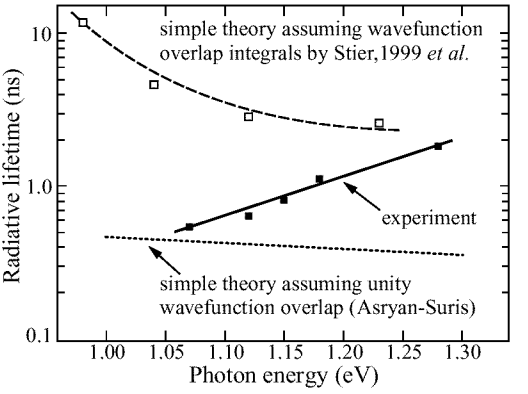

Fig. 2. Radiative lifetime of an isolated QD as a function of the photon energy

in InAs QDs having different size (pyramid base length between 10 and 20 nm) derived at low temperatures using both resonant and non-resonant excitation is shown in the figure by solid squares. One can see that these results agree only in the case of smaller QDs, where the role of light confinement on radiative lifetime seems to be relatively weak. As opposite, as the QD size increases, the theoretical dependence and experimental values differ qualitatively. As the structural quality and luminescence efficiency of larger QDs remains high allowing high-efficiency high- power device applications [15], the reason for such a discrepancy can be only related to the discussed light confinement effect at QD.

3. Conclusion

We have introduced a concept of light confinement and investigated its role in electromagnetic response of QDs. We calculated significant diffraction-induced shift of the main QD exciton line. Evaluation of the radiative lifetime for spherical QD and its correlation to the QW radiative lifetime shows the origin of the fascinating light-amplifying properties of QDs as compared to QWs and creates a basis for solving of a large number of electrodynamic problems of QDs and QD ensembles. We show that the experimentally measured radiative lifetime qualitatively disagrees with theoretical predictions arising from models neglecting the light confinement effect at QD. Thus one needs to consider redistribution of the electromagnetic wave caused by QD to reach optimized device geometry. This is particularly true for a QD inserted in a microcavity, where cavity modes may interfere with intrinsic photon modes of a single QD. In our paper we mainly restricted ourselves to the spherical model of QD. Different QD configurations like disks or pyramids can be investigated using direct computation on the basis of the well-developed method of classical electrodynamics.

Acknowledgments

The research is partially supported through INTAS under project 96-0467 and the NATO Science for Peace Program under project SfP-972614. N.N.L. acknowledges DAAD Professorship. We are grateful to L. Asryan for helpful discussions.

References

[1] D. Bimberg, M. Grundmann and N. N. Ledentsov, *Quantum Dot Heterostructures* (John Wiley & Sons, Chichester, 1999).
[2] S. Schmitt-Rink, D. A. B. Miller and D. S. Chemla, *Phys. Rev. B* **35**, 8113 (1987).
[3] O. J. F. Martin, C. Girard and A. Dereux, *Phys. Rev. Lett.* **74**, 526 (1995).
[4] B. Hanewinkel *et al.*, *Phys. Rev. B* **55**, 13715 (1997).
[5] G. Ya. Slepyan *et al.*, *Phys. Rev. B* **59**, 12275 (1999).
[6] S. A. Maksimenko *et al.*, *J. Electron. Mater.* (2000, to be published).
[7] D. Gammon *et al.*, *Phys. Rev. Lett.* **76**, 3005 (1996); *Science* **273**, 87 (1996).
[8] S. A. Maksimenko *et al.*, *Semicond. Sci. Technol.*, (2000, to be published).
[9] L. D. Landau and E. M. Lifshitz, *Electrodynamics of continuous media* (Pergamon Press, Oxford, 1960).
[10] M. Asada, Y. Miyamoto and Y. Suematsu, *IEEE J. Quant. Electr.* **22**, 1915 (1986).
[11] N. A. Khiznjak *Integral equations of macroscopic electrodynamics* (Naukova Dumka, Kiev, 1986, in Russian).
[12] F. Tassone, F. Bassani and L. C. Andreani, *Phys. Rev. B* **45**, 6023 (1992).
[13] L. V. Asryan and R. A. Suris, *Semicond. Sci. Technol.* **11**, 554 (1996).
[14] O. Strier, M. Grundmann and D. Bimberg, *Phys. Rev. B* **59**, 5688 (1999).
[15] A. E. Zhukov *et al.*, *IEEE Photonics Technol. Lett.* **13**, 1345 (1999).